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Fabrication of tantalum oxyfluoride and oxynitride thin films via ammonolysis of sol–gel processed tetraethoxo (β -diketonato) tantalum (V) precursors for enhanced photocatalytic activity

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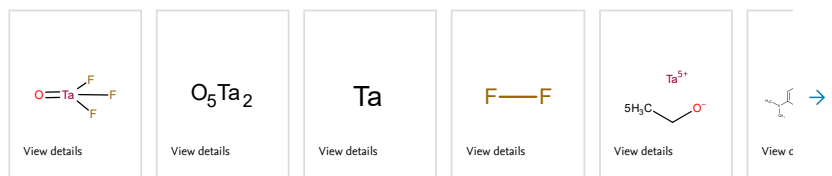
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Abstract

In the present report, the generation of Tantalum oxyfluoride and oxynitride upon ammonolysis of the gel obtained from modified tantalum-alkoxo complexes is reported. To the best of our knowledge, this is the first report of the formation of tantalum oxyfluoride thin films via ammonolysis of the β -diketone modified tantalum-alkoxo complex $[\text{Ta}(\text{OEt})_4(\text{CF}_3\text{COCH}_2\text{COCH}_3)]_m$. The integration of nitrogen and fluorine in lattice sites of metal oxides leads to significant reduction in the bandgap, resulting in their activation under visible light. Moreover, in this report the effect of the modified alkoxide precursors and ammonolysis on the photophysical properties of Ta_2O_5 thin films have also been investigated and compared with the results obtained from films fabricated from unmodified tantalum (V) ethoxide. ^1H NMR, ^{13}C NMR and elemental analyses confirmed successful modification of tantalum (V) ethoxide to $[\text{Ta}(\text{OCH}_2\text{CH}_3)_4(\text{CH}_3\text{COCHClCOCH}_3)]_m$ (1), $[\text{Ta}(\text{OCH}_2\text{CH}_3)_4(\text{CF}_3\text{COCH}_2\text{COCH}_3)]_m$ (2) and $[\text{Ta}(\text{OCH}_2\text{CH}_3)_4(\text{CH}_3\text{COC}(\text{CH}_3)_2\text{COCH}_3)]_m$ (3). The fabrication of Ta_2O_5 thin films involved the spin casting of the gels of modified tantalum alkoxo complexes (processed by sol–gel method) on to glass substrate. X-ray photoelectron spectroscopy results show that nitrogen was incorporated into the ammonolyzed films fabricated from complex precursors (1) and (3), while the presence of fluorine as tantalum oxyfluoride was confirmed in the ammonolyzed film fabricated from complex (2) precursor. The optical characterization insinuate bandgap narrowing from 3.55 eV for undoped film prepared from tantalum (V) ethoxide to 3.47 eV for undoped film prepared from $[\text{Ta}(\text{OEt})_4(\text{CF}_3\text{COCH}_2\text{COCH}_3)]_m$ and 3.05 eV for ammonolyzed film obtained from $[\text{Ta}(\text{OEt})_4(\text{CF}_3\text{COCH}_2\text{COCH}_3)]_m$ precursor. Furthermore, enhanced photocatalytic efficiency of the films is demonstrated by degradation of methylene blue dye. © 2021, The Author(s), under exclusive licence to Springer Science+Business Media, LLC, part of Springer Nature.

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